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TEMPORARY BEHAVIOR OF PROCESSES IN THE  
DIELECTRIC POLARIZATION OF BARIUM TITANATE AND ROCHELLE SALT

BY

N. S. NOVOSIL'TSEV

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Very large values of the dielectric constant of barium titanate are observed in the presence of Rochelle salt.

N. S. Novosil'tsev

Study of the properties of a new ferroelectric, barium titanate, discovered by B. M. Vul, shows that the mechanism of the processes, which explains its high dielectric constant, is essentially similar to that which takes place during the polarization of classical ferroelectrics typified by Rochelle Salt. This is especially noticeable in the observed duration of such processes. In the present paper there is given a comparison of the duration of the discharge curves for samples of barium titanate and Rochelle salt along the A axes, at various temperatures.

Judging by recent results the ferroelectric properties of barium titanate are very sensitive to impurities. This leads to the idea that here we have to do with disturbances which produce changes in the crystal lattice.

In the ordinary methods of studying the dielectric constant by means of capacity measurements, using the resonance method at various frequencies and the ballistic method at constant fields, the results are characteristic of a substance undergoing on the whole a cycle of processes.

For an explanation of the mechanism depending on the greater susceptibility of ferroelectrics, it is of interest to know in detail how the time processes of dielectric polarization take place. Goldecke,<sup>1</sup> by means of a rather cumbersome vacuum tube circuit, made a similar study of this type on the processes of depolarization in Rochelle salt. Goldecke's circuit allowed him to observe indirectly such processes in discrete moments of time.

In the present work a specially constructed oscillograph was used to directly produce an uninterrupted record of the processes of discharge of a capacity, with the dielectrics under study, for the active resistance.

By connecting the samples directly to the deflector plate, (and) by-passing the ordinary stages of the oscillograph circuit, it was certain that the (resulting) curves reflect precisely the time constant for the samples and not for the circuit.

A functional scale on plexiglas was used for deciphering the oscillogram. The sequence of operations in the treatment of the observations on the plane eliminated the influence of the distortions of the oscillograms (resulting) from the projection on the convex screen of the tube.

In order to check the reliability of the operation of all the connections to the apparatus and the working conditions, the oscillogram values of the dielectric constant of the barium titanate sample were calculated from the slope of the lines on the final graphs (see Figs. 1b, 2b and the straight line section on Fig. 3b) at various temperatures in the range  $48^{\circ}$  to  $164^{\circ}$ . The curve drawn through the calculated points, reproduces in detail the path corresponding to the curves as plotted from data obtained from the ordinary resonance method. The values of the dielectric constant of barium titanate resulting from adjustment with the oscillogram are presented in the table as a function of temperature

On Figs. 1a, 2a and 3a there are shown the curves of the discharge of a sample of a ceramic condenser of barium titanate in the form of a disc 12 mm in diameter and 2 mm thick for  $R = 12 \text{ Mw}$ . The curves are plotted from points taken from the oscillogram, as a rule every  $1 \times 10^{-3} \text{ sec}$ . There was no need to take points much closer than this. On the vertical axes there is plotted in mm. the corrected lengths corresponding to the ordinates of the oscillogram on a flat screen 20 x 30 cm where 1 mm corresponds approximately to 1.5 volt potential difference on the condenser under study.

(see table on bottom of pg. 810)

$t^{\circ}$  ----  
 $\epsilon \times 10^{-2}$  ----

The curves on Fig. 1a refer to temperatures below the Curie point and those of Fig. 2a above the Curie point. Separate on Fig. 3a are the curves for temperatures much above the Curie point, where the leakage and self-discharge of the sample become appreciable. The curves of the discharge of the sample by itself in this case are plotted on the upper part of Fig. 3a. As will be shown below, in this case the curves of discharge of the sample on an external resistance also begins to have substantial peculiarities. The

curves above and below the Curie point as presented on Figs. 1a and 2a, fully conform to the equation

$$V_t = V_0 e^{-t/RC}$$

In fact, as shown in the graphs (Figs. 1b & 2b) the relations  $\ln V_0/V_t = \frac{1}{RC} t$  are quite linear and proceed strictly from the co-ordinate origins. This means, that directly from the moment of the origin of the discharge the barium titanate condenser already exhibits a large dielectric constant. Then the polarization mechanism deteriorates very quickly, and the dielectric constant does not vary during the process of discharge. Precisely for this reason barium titanate in contrast to Rochelle salt does not have a lower dielectric constant even at very high frequencies as was shown by direct measurements.<sup>2,3</sup>

Only considerably above the Curie point do the curved lines on Fig. 3 speak for the appearance of a somehow or other retarded active mechanism of polarization. In all probability, this is an inner layer high voltage polarization which develops, as was shown by Kosman<sup>4</sup> for a large number of solid bodies at sufficiently high temperatures. That this in fact has taken place in the present case might be inferred from the oscillogram of the discharge of the sample by itself. Such oscillograms were taken down for all temperatures simultaneously with the discharge curves on the external resistance. However, above and below the Curie point at temperatures as shown on the graphs of Figs. 1a, 2a and 1b and 2b, for a time of 0.02 sec, no signs of a self-discharge have appeared on the oscillograph.

As shown by the measurements of Hodakov on  $\tan \delta$ , barium titanate at the Curie point undergoes some lowering. This is probably the case from the point of view discussed, as obtained from the oscillograph. The capacity at the Curie point increases, and nothing unusual happens since there is loss during the Curie point transition.

Only at sufficiently high temperatures far from the Curie point does the self-discharge shown on the oscillograph set in to produce a drift of the discharges through the samples which gradually develops during further rise in temperature. This also creates the conditions necessary for the formation of an inner layered polarization. The character of the phenomenon in Rochelle salt is quite different.

On Fig. 4 is plotted a logarithmic graph constructed from the oscillogram discharge for a resistance of 1.45 M ohms for a sample of Rochelle salt 40 x 40 x 6 mm, cut from a single crystal perpendicular to the single ferroelectric axis. Exploration in the field of temperatures in the region above the Curie point showed that above it beyond the region of spontaneous polarization the line is straight and passes through the origin of the co-ordinates. Here Rochelle salt shows itself to be a dielectric with susceptibility unchanged in the process of discharge and with negligible losses. On the other side of the Curie point a bending of the line is at once produced, and this speaks for a delayed action of the mechanism of polarization. This is explained when one observes the decrease in susceptibility of Rochelle salt at increased frequencies.

Barium titanate also has a logarithmic graph which is straight at the beginning and then proceeds from the co-ordinate origins. The noticeable bend in the curve only becomes apparent after a comparatively large interval of time (greater than  $5 \times 10^{-3}$  sec). This indicates a second additional mechanism of polarization. It is possible to show that this mechanism will make itself felt only at sufficiently low frequencies, such as the case if the condenser should be periodically recharged in one direction, as in a filter circuit.

The graph for Rochelle salt is curved at the beginning of the discharge and straight at the end. Prolongation backward of the rectilinear section of these graphs does not hit the co-ordinate origins. This indicates a comparatively larger relaxation time for the polarization mechanism in Rochelle salt than that of barium titanate.

In this way it is once more confirmed that the ferroelectric properties in barium titanate are essentially different from those that are found in Rochelle salt and similar dielectrics.

In conclusion I wish to thank Prof. B. M. Vul for his interest in the work and for discussions of the results.

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Physics-Mathematical Institute  
Rostov State University

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C. A. BIELING

J. R. FISHER